

Abstract No. duff89

Characterization of Sorbed Neptunium on Monosodium Titanate

M.C. Duff, D.B. Hunter, D.T. Hobbs, M.J. Barnes and S.D. Fink (Westinghouse Savannah River Company)
Beamline(s) X23A2

Introduction: The current preferred design for the Salt Processing Facility at the Savannah River Site includes use of monosodium titanate (MST) to remove radiostrontium and actinides from the radioactive High Level Waste (HLW) solutions. Little is known about the structure of MST but it most likely contains Ti^{4+} octahedra as opposed to tetrahedral Ti^{4+} . Moreover it is not known why MST has a strong affinity for actinides such as Np in alkaline (pH 14) HLW simulant solutions that are rich in Na^+ ion.

Methods and Materials: To study the chemistry of sorbed Np on MST in HLW salt simulant solutions we used extended X-ray absorption fine structure spectroscopy (EXAFS), X-ray absorption near-edge structure (XANES) spectroscopy and molecular modeling techniques. Neptunium-loaded MST solids were prepared in HLW salt simulant solutions to have Np loadings ranging between 1000 to $\sim 10,000 \text{ mg Np kg}^{-1} \text{ MST}$. The Np was added to the HLW simulant solutions from acidic stock solutions of Np(V) and of Np(IV). Data collection was conducted at the L_3 absorption edge of Np using a 5-grid Lytle fluorescence detector.

Results: The Np-XANES studies indicate all of these sorbed Np species had the same average oxidation state which was +IV or +V. The EXAFS analyses indicate that the sorbed Np [added to the HLW simulant as Np(V)] exists with several first shell Np-O interactions. Higher shell Ti and Np atoms were also observed. We conclude that Np added as Np(V) at high loadings, sorbs as polymeric species via an inner sphere adsorption. Samples with lower Np loadings added as Np(V), were somewhat similar except no second shell Ti was observed. Analyses for sorbed Np added initially to the HLW simulant as Np(IV), do not have higher shell Ti or Np, which indicates that the Np in this sample is monomeric and exhibits outer sphere sorption. These preliminary results indicate that both oxidation state and loading levels effect the binding mechanism of Np to MST and that further studies are warranted. WSRC-MS-2001-00725.